Composition Change in Binary-component Spray Vaporization at Atmospheric Pressure

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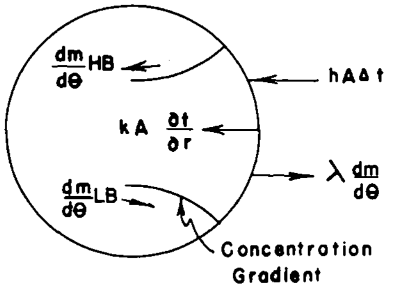
This paper describes an investigation of the factors affecting the change in composition with vaporization of a binary-component spray in heated air at atmospheric pressure. The behavior of the system orthodichlorobenzene-tetrachloroethylene was studied in air ranging from 400° to 1,000°F. Initial drop diameters in the sprays were in the 20- to 400-μ range. The experimental data obtained indicated that the rate of change of spray composition during vaporization was affected only by the chamber-air temperature, the initial feed composition, and the nozzle characteristics.

Equations are presented which mathematically describe the vaporization process, liquid diffusion being assumed within the drop controls. These equations have been solved by a stepwise procedure for three initial drop sizes. Results of these calculations have been added statistically according to the initial-drop-size distribution in order to predict the vaporization behavior of the spray. Details of this process of statistical combination have been summarized by Culverwell (*1*).

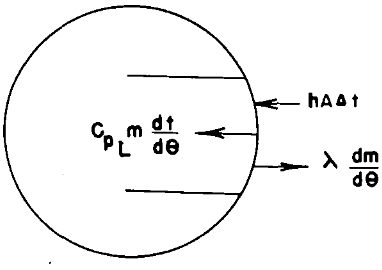
The calculations agreed with the experimental data previously obtained during the first 5 in. of nozzle-to-tray travel. After 5 in. the deviation was great. Future experimental work to determine more precise values of drop-size distribution, initial drop velocity, and liquid diffusion coefficient may result in more complete agreement.

A principal problem encountered in the design of jet-engine-fuel-injection systems is the prediction of fuel-vaporization behavior. In this problem atomization, vaporization, and combustion are interrelated, with atomization influencing vaporization, which in turn affects combustion of the fuel. Other processes also require a knowledge of the mechanism of spray vaporization for a good engineering design. These include spray drying and humidification operations in particular. It seems that the development of a method of predicting the behavior of multicomponent sprays with respect to composition change during vaporization would have considerable practical value.

The mechanism of mass transfer in a binary droplet may be bracketed by the two extremes of liquid diffusion controlling and gas diffusion controlling as shown in Figure 1. The droplet temperature is such that the heat transfer to the drop from the air equals the heat required by mass transfer from the drop and heat conduction into the drop. As the air temperature is increased, the droplet temperature approaches the boiling point. When the air temperature is increased beyond the point where the drop is at its boiling point, the mechanism may change in the direction of constant composition vaporization, owing to the comparatively slow rate of liquid mass transfer within the droplet.



LIQUID DIFFUSION CONTROLLING



GAS DIFFUSION

Fig. 1. Mass and heat transfer in a binary-component drop with liquid diffusion controlling and with gas diffusion controlling.

The investigation described in this paper consisted of the measurement of the variables affecting the vaporization of a binary spray, followed by the development of a prediction method from basic mass and heat transfer relationships. The variables studied for their effect on the composition change–vaporization relationship were air temperature, feed composition, feed temperature, feed pressure, and distance from nozzle. Measurements were made with a typical spray composed of a large range of drop sizes, while the mechanism calculations were made for one particular initial drop size at a time. Then the bahavior of the whole spray was synthesized by summing the behavior of the various individual drop sizes according to the drop-size distribution of the spray.

REVIEW OF PREVIOUS WORK

Previous work on droplet vaporization has been carried out primarily on single drops of pure materials. There has been some work with sprays, but none of this has considered the change of composition during the vaporization of a multicomponent spray.

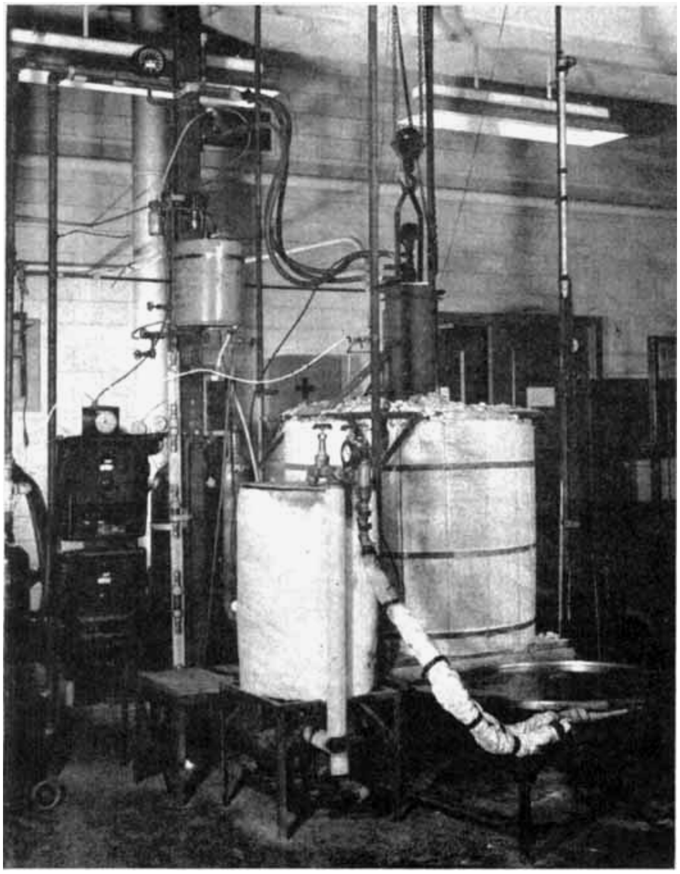


Fig. 2. Photograph of movable nozzle-spray chamber showing feed system and ice-water–cooled spray collection tray.

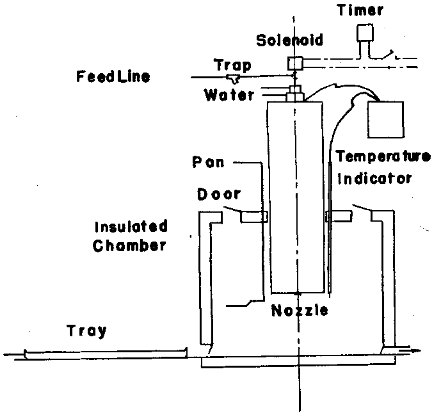


Fig. 3. Schematic sketch of spray chamber.

Lamb and O'Brien (*5*) computed the change in composition of a four-component fuel drop, assuming ideal-solution behavior. The vaporization mechanisms compared were (1) true-boiling-point vaporization, (2) differential vaporization, (3) equilibrium flash vaporization, (4) A.S.T.M. vaporization, and (5) vaporization predicted by gas diffusion. These authors suggested that experimental work should be carried out on common fuel systems in order to determine which of these possible mechanisms was most likely to be the true one.

Selden and Spencer (*10*) carried out preliminary experiments designed to indicate the vaporization mechanism. They reported that vaporization began immediately upon injection of the fuel and that the initial heat transfer rate was a function of the initial air-to-drop temperature difference. Godsave (*3*) distinguished between two possible mechanisms which might control vaporization rate: (1) when the air temperature is below the drop boiling point, gas diffusion controls the vaporization rate; and (2) when the air temperature is considerably above the drop boiling point, the heat transfer rate *controls vaporization.*

Topps (*11*) confirmed the second mechanism in his work with single drops of pure material in the 300- to 500-μ-diam. range falling at terminal velocities through air up to 630°C. He concluded that heat was conducted into the drop, as indicated by radius increase during the early stages of the evaporation. For air temperatures 200°C. or more above the drop boiling point, the wet-bulb temperature was considered to be the boiling point, which is a good approximation. In addition, Topps concluded that multicomponent fuels evaporated as pure materials as far as vaporization rate was concerned. Unfortunately, the composition change accompanying the vaporization was not determined.

Ranz and Marshall (*9*) developed heat and mass transfer correlations for droplet vaporization based on experiments made with single water drops from 600- to 1,100-μ diam. in air streams up to 220°C. at Reynolds numbers of 200. In this work a 0.5-mil thermocouple at the drop center indicated the equilibrium drop temperature to be at the wet-bulb temperature. This wet-bulb temperature was also determined by Ingebo (*4*) in correlating data on the vaporization of pure liquids from a 0.688-cm.-diam. cork ball in air streams up to 500°C. El Wakil, Uyehara, and Meyers (*2*) developed an incremental-trial-and-error method for prediction of the vaporization of pure droplets, assuming mass transfer from the drop controlled by gas diffusion.

EXPERIMENTAL EQUIPMENT AND TECHNIQUE

The spray unit shown in Figures 2 and 3 consisted of (1) feed system, (2) heating chamber, and (3) spray collection system. The feed line consisted of a feed reservoir and a system of glass volume gauges with a valve arrangement so that nitrogen pressure on the feed line might be applied during spraying and released afterward. The feed line was connected to the movable nozzle and feed-line cooling assembly by a flexible metallic hose. The feed cooling line was a triple-pipe heat exchanger with cooling water entering the inner annulus at the top and passing to the outer annulus at the bottom of the exchanger. Flexible hoses were also used on the water-coolant lines. The feed nozzle was screwed into a coupling at the bottom of the feed cooling exchanger and the valve seat was located in this coupling, to which a thermocouple was attached $1\frac{3}{8}$ in. above the nozzle face. The valve stem extended from this valve seat to the solenoid located on the top of the exchanger at the feed inlet. The solenoid valve and a timer were operated simultaneously through a single switch. The feed exchanger was housed in an 8-in.-diam. sheet-iron shell so insulated that the feed cooling would not appreciably affect the chamber temperature. The entire housing was movable by a chain hoist. The nozzle face was flush with the bottom of this insulated shell and a movable thermocouple permitted measurement of the air temperature at the nozzle height. A movable drip pan was located so that it might be swung under the nozzle to catch the drips after the spray was turned off after each run. The chamber was heated by nichrome resistance wires wound around the outside. The chamber air was exhausted at the end of each run by a blower which caused a flow of air through the chamber from top to bottom.

The spray was collected on an ice-water–cooled internally baffled circular tray, which could be slid into the chamber through a door at the bottom of the cham-

Table 1. Typical Experimental Data

67% by Volume o—$C_6H_4C._2$ in C_2Cl_4
Nozzle ¼ LN No. 6
Feed pressure 30 lb./sq. in. gauge
Tray correction 3 cc.
Exit tray coolant 40°F.

Temperature, °F.			Spray time, sec.	Tray distance, in.	Residual composition			Volume, cc.					Volume % vaporized
Chamber	Nozzle	Tray			Refractometer Temp., °C.	Refractometer Index	Volume % H. B.	Reading Start	Reading End	Feed	Collected	*Corrected* vaporized	
412	75	30	18.57	15	20.0	1.5382	71.8	0.5	63.5	63.0	35.5	24.5	38.8
413			40.21			1.5389	73.2	0	132.5	132.5	82.0	47.5	35.9
416			28.23			1.5391	73.6	133.5	223.5	90.0	57.0	30.0	33.3
402			51.29			1.5394	74.3	62.0	225.0	163.0	108.0	52.0	31.9
403			68.84			1.5394	74.3	0	224.5	224.5	146.0	75.5	33.7
408			18.16	10	19.5	1.5373	69.7	2.5	63.5	61.0	44.0	14.0	23.0
400			39.99			1.5372	69.5	1.5	133.0	131.5	102.0	26.5	20.1
407			28.25			1.5380	71.5	132.5	223.5	90.5	71.0	16.5	18.3
410			50.00			1.5380	71.5	63.5	223.5	160.0	129.0	28.0	17.5
410			66.29			1.5380	71.5	−4.0	223.0	219.0	176.0	40.0	18.3
390			18.93	5	21.5	1.5356	67.2	−1.0	63.0	64.0	54.5	6.5	10.2
398			49.59			1.5362	68.5	62.0	222.5	160.5	144.0	13.5	8.4
395			39.22			1.5362	68.5	3.0	132.5	129.5	114.5	12.0	9.3
394			28.10			1.5360	68.0	133.0	223.5	90.5	84.0	3.5	3.9
410			66.87			1.5362	68.5	5.0	273.0	218.0	197.0	18.0	8.3
590	80	30	27.17	5	27.0	1.5365	73.3	136.5	225.0	88.5	—	—	—
596			16.75			1.5360	72.5	4.5	62.0	57.5	46.0	8.5	14.8
585			17.89			1.5364	73.3	0.5	61.5	61.0	49.0	9.0	14.8
595			50.49			1.5371	75.0	59.0	224.0	165.0	137.0	25.0	15.2
595			21.58			1.5367	74.0	60.0	131.0	71.0	60.0	8.0	11.3
585			66.67			1.5374	75.5	4.0	222.5	218.5	179.0	36.0	16.5
575			39.64			1.5368	74.0	3.0	131.5	128.5	108.5	17.0	13.2
607			16.97	10	26.2	1.5379	75.8	3.0	63.0	60.0	36.5	20.5	34.2
606			44.24			1.5386	77.0	66.0	224.0	158.0	99.0	56.0	35.4
597			36.48			1.5384	77.0	3.5	133.0	129.5	79.5	47.0	36.3
610			36.79		26.7	1.5398	79.7	4.5	133.0	128.5	52.5	73.0	56.8
785	50	30	41.03	15	21.0	1.5442	85.0	0	131.5	131.5	44.0	84.5	64.3
790			27.62			1.5438	84.8	133.0	223.0	90.0	30.0	57.0	63.4
804			63.19			1.5453	88.5	2.0	222.0	220.0	70.5	146.5	66.5
783			17.11			1.5444	86.5	3.0	61.0	58.0	17.5	37.5	64.6
795			50.46			1.5442	85.0	64.0	222.5	158.5	54.0	101.5	64.0
802			38.88	10		1.5430	83.0	3.0	131.5	129.5	70.0	56.5	43.6
805			27.71			1.5420	81.0	134.0	222.5	88.5	50.0	35.5	40.0
810			17.23			1.5425	82.5	3.5	61.5	58.0	29.5	25.5	44.0
805			49.89			1.5433	84.0	62.0	222.5	160.5	85.0	72.5	45.2
1002	55	30	16.23	10	23.0	1.5427	84.0			50.0	21.0	26.0	52.0
1008			10.37			1.5438	86.5			32.0	14.5	14.5	45.3
1009			21.39			1.5428	84.5			65.0	30.0	32.0	49.2

ber. Liquid was removed from the tray by squeegee and vacuum line into a graduated cylinder. The tray coolant was pumped to the tray from an ice-water reservoir. Vaporization and condensation on the tray were checked under the conditions of operation and were found to be negligible. Spraying Systems nozzle ¼ LN 4 was used throughout.

EXPERIMENTAL DATA

The experimental procedure used was designed to give data which would permit the construction of a vaporization-composition diagram for the system being studied, 67% (by volume) orthodichlorobenzene and 33% tetrachloroethylene. Measurements were made at given nozzle-to-collection-tray distances under given fir and feed conditions to determine, as a aunction of the volume of feed input, the composition of the unvaporized spray collected on the tray and the fraction of the input feed vaporized.

Refractive index was measured to determine the composition of the residual liquid on the tray as mole fraction of high boiler. The high boiler present is the main problem in the "tail-end volatility" of fuel, and for this reason compositions were measured in terms of the high boiler. The fraction of feed vaporized was secured by difference between the measured value of feed sprayed and the measured volume of unvaporized spray collected on the cooled tray. Complete data for the system studied are presented in Table 1, for 400°, 600°, and 800°F. with preliminary data* at 1,000°F. The tray correction of 3 cc. represents the volume of liquid which was not recoverable from the tray owing to sticking of liquid to the tray surfaces. Details of this correction and other calibration runs are summarized by Culverwell (*1*).

The experimental runs, for which data are given in Table 1, were all unsteady state in nature, starting with the chamber full of pure air at the particular temperature indicated. However, once the run got underway the temperature and composition of the air-vapor mixture in the chamber varied in a very complex manner. In order to secure data at known conditions, the measurements made at different feed-input volumes were extrapolated to zero feed volume. At zero feed volume, the chamber air temperature and composition were at their known initial values. Data obtained by such an extrapolation represented the behavior of the first few droplets of spray as they fell through an unchanging environment similar to that in existence throughout the entire chamber at the instant before spraying began. Hence, original data taken from a series of unsteady state runs were combined to yield data which described the result of a hypothetical steady state operation. A typical set of extrapolations is shown in Figure 4. Linear extrapolations were used, which seems to be a reasonable approximation. Other such extrapolations were also made, as summarized by Culverwell (*1*).

Extrapolated values of residual composition in mole-fraction high boiler were

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*Tabular material has been deposited as document 5054 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or 35-mm. microfilm.

plotted against extrapolated values of mole-fraction feed vaporized to give the curves in Figure 5. Three of the points in Figure 5 come from the extrapolations shown in Figure 4. Similar extrapolations of additional data were required for the rest of the points shown. Figure 5 indicates that the shape and location of the vaporization-composition curve are affected primarily by air temperature, feed composition, and nozzle size. Other variables, such as feed temperature, feed pressure, and nozzle-to-tray distance locate a point on the curve but do not change the curve shape.

DEVELOPMENT OF MECHANISM EQUATIONS

The following is a brief summary of the procedure used for the development of the mathematical relationships existing under the assumption of a particular mechanism of droplet vaporization, that of liquid diffusion controlling. The equations were developed to describe the behavior of a single drop, after which they were combined statistically to predict the behavior of a spray comprised of a wide range of droplet sizes.

In the following mathematical development it was assumed that (1) convection within the drop was negligible, (2) mass transfer of each component in the drop was controlled by liquid diffusion, (3) heat transfer within the drop was controlled by thermal conduction, (4) the surface composition of the drop was 100% high boiler, and (5) the surface temperature of the drop is the boiling point (of the high boiler). To make the resultant equations possible of solution, they were set up to give expressions for the drop radius, drop temperature, drop composition, and drop velocity as a function of the distance traveled from the nozzle, with the distance expressed in terms of an increment of travel, $L_2 - L_1$. Thus a stepwise calculation procedure was required, moving from increment to increment and repeating the computations at each increment, a tedious process at best. Details of the equation derivations and of their use in computation are to be found in the original work upon which this paper is based (*1*).

By means of a heat balance around a single drop combined with a low-boiler mass balance around the drop, the following equation for r_2/r_1 may be developed:

$$\ln \frac{r_2}{r_1} = -\frac{1}{\lambda_{HB}} \cdot \left[\left(\frac{h \Delta t}{r u \rho_L} \right)_m (L_2 - L_1) - \frac{M_{LB}}{3M} \cdot (\lambda_{HB} - \lambda_{LB})(X_{m_2} - X_{m_1}) - \frac{C_{PLm}}{3} (t_{m_2} - t_{m_1}) \right] \quad (1)$$

Quantities written with a subscript *m* should be evaluated at the appropriate volume mean, which was assumed to be that at a point within the drop where $r = 0.794 r_{max}$.

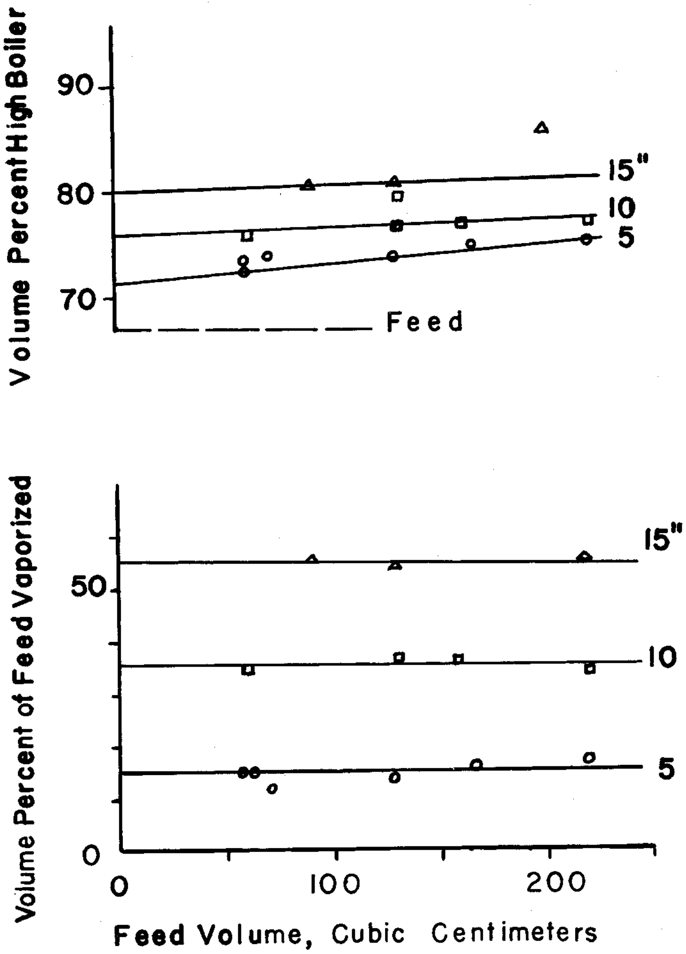

Fig. 4. Extrapolation to zero feed volume of residual composition and volume percentage vaporized for 67%-by-volume orthodichlorobenzene in tetrachloroethylene sprayed at 15°C. and 30 lb./sq. in. gauge into 316°C. air.

For the next relationship, a thermal conduction heat balance around a differential volume of the drop is required, a spherical drop and radially symmetrical temperature gradients within the drop being assumed. The result is the following equation for $t_{m_2} - t_{m_1}$:

$$\frac{t_{m_2} - t_{m_1}}{t_b - t_0} = 0.404 \left[\exp\left\{ \frac{-2.465 \alpha_{L_1} L_1}{u_m {r_1}^2} \right\} - \exp\left\{ \frac{-2.465 \alpha_{L_2} L_2}{u_m {r_1}^2} \right\} + \cdots \right] \quad (2)$$

On the basis of liquid diffusion within the droplet by means of a diffusional mass transfer balance over a differential element inside the drop, the following relationship between X_{m_1} and X_{m_2} may be developed:

$$\frac{\frac{\rho_{L_2}}{M_2} X_{m_2} - \frac{\rho_{L_1}}{M_1} X_{m_1}}{\frac{\rho_{HB}}{M_{HB}} t_{HB} - \frac{\rho_{L_0}}{M_0} X_0} = 0.404 \left[\exp\left\{ \frac{-2.465 D_{HB_1} L_1}{u_m {r_1}^2} \right\} - \exp\left\{ \frac{-2.465 D_{HB_2} L_2}{u_m {r_1}^2} \right\} + \cdots \right] \quad (3)$$

Finally drag relationships for the two-dimensional motion of a spherical particle were applied to give the following equations for the components of the droplet velocity:

$${u_{v_2}}^2 = 2.66 \frac{r_1 g \rho_{L_1}}{f_1 \rho_a} + \left[{u_{v_1}}^2 - 2.66 \frac{r_1 g \rho_{L_1}}{f_1 \rho_a} \cdot \exp\left\{ -0.75 \frac{f_1 \rho_a}{r_1 \rho_L} \right\} (L_{v_2} - L_{v_1}) \quad (4)$$

$$U_{h_2} = U_{h_1} \exp\left\{ \frac{-2.66 f_1 \rho_a}{r_1 \rho_{L_1}} \right\} (L_{h_2} - L_{h_1}) \quad (5)$$

In the application of the foregoing relationships [Equations (1) through (5)] a good deal of supplementary information was required. Physical properties of the system being studied were secured from

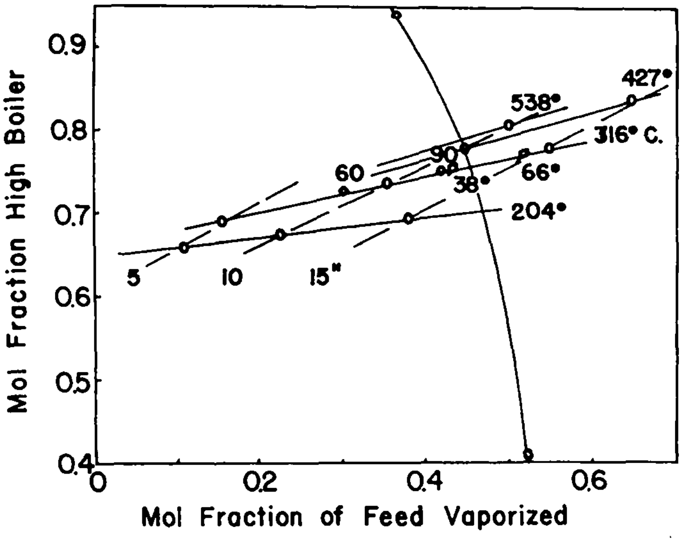

Fig. 5. Composition-vaporization diagram for orthodichlorobenzene-tetrachloroethylene.

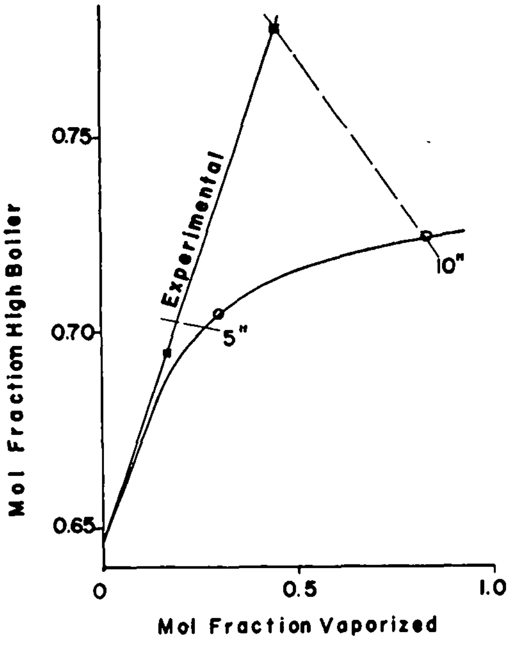

Fig. 6. Comparison of calculation with experimental data on vaporization-composition diagram for 67%-by-volume orthodichlorobenzene in tetrachloroethylene at 15°C. and 30 lb./sq. in. gauge sprayed through Spraying Systems nozzle ¼ L.N. 4 into 427°C. air.

the literature or were measured in the laboratory. Liquid diffusion coefficients for orthodichlorobenzene in tetrachloroethylene were computed by the method of Wilke (*12*) and the correction for nonideal solutions proposed by Powell, Roseveare, and Eyring (*8*) with activity coefficients computed by means of a Van Laar three-suffix equation using experimentally determined boiling-point data. The heat transfer coefficient was computed by the correlation of Ranz and Marshall (*9*).

CALCULATION PROCEDURE AND RESULTS

In order to compute spray vaporization and composition change predicted by the equations previously summarized, three initial drop sizes were selected and computations made in a stepwise manner, by use of 2-cm. increments along the drop trajectory. The calculated values secured at a given nozzle-to-tray distance were then added statistically in accordance with the initial drop-size distribution of the spray in order to produce a typical value for the bulk spray.

The method of solution is summarized in detail by Culverwell (*1*). For each increment of each drop size, calculations were made according to the following plan:

1. Initial drop-size distribution was measured experimentally by use of a slide-sampling and photographic-counting procedure. The data are summarized by Culverwell (*1*).
2. Initial drop velocities were secured from the data of Landsbaum (*6*) for water, corrected for the binary being used. The following values for initial velocity resulted:

d_0, μ	u_0, cm./sec.
200	1,450
300	1,120
400	937

3. Physical properties were obtained from Perry (*7*). Supplemental data were obtained in the laboratory.
4. The vertical component of velocity at the end of one increment of trajectory travel was calculated by use of Equation (4) with $L_{v_2} - L_{v_1} = 2$ cm.
5. The horizontal component of velocity was calculated from Equation (5), after which the new trajectory angle and the resultant velocity were computed.
6. The mean drop temperature was next calculated by means of Equation (2).
7. By the Wilke (*12*) method the liquid diffusion coefficient was computed.
8. Equation (3) was used to calculate the mean drop composition at the end of the increment.
9. By means of the Ranz and Marshall (*9*) correlation, the heat transfer coefficient was computed.
10. Finally the new drop radius was calculated from Equation (1).
11. Steps 4 through 10 were repeated for additional 2-cm. increments, until the desired nozzle-to-tray distances were obtained.

At nozzle-to-tray distances of 2, 5, and 10 in. the calculated results were added statistically according to the initial drop-size distribution as measured previously. The resulting composition vs. mole-fraction vaporized plot is shown in Figure 6, with the experimental results for the same initial conditions shown for comparison. The calculated results agree rather well for the first five inches of fall, after which considerable deviation is indicated. Perhaps actual measurement of diffusion coefficients or better data on drop sizes and velocities may result in greater agreement between calculated and experimental results at distances greater than 5 in. Also it is possible that a substantial portion of the heat may be transferred to the drops by a radiation process, the effect of which increases as the droplets fall from the nozzle to the tray.

CONCLUSIONS

The experimental data, when extrapolated to zero feed volume and plotted on a vaporization-composition diagram like Figure 5, indicate the validity of the following generalizations:

1. The change of residual spray composition with vaporization is a function of initial composition, chamber air temperature, and nozzle characteristics for the particular binary, in this case orthodichlorobenzene-tetrachloroethylene.
2. The other variables investigated—feed pressure, feed temperature, and nozzle-to-tray distance—have the effect of locating the position on the vaporization-composition curve but do not affect the shape of the curve.

The calculated results for the binary based on the assumption of liquid-diffusion-controlling mass transfer indicate the smaller droplets slowing down, increasing in temperature, changing in composition, and decreasing in radius more rapidly than the larger drops. Such behavior is consistent with experience. Comparison of the calculated data with experimental results as shown on a vaporization-composition diagram indicates agreement during the first five inches of nozzle-to-tray height. It appears that a thermal conduction and liquid diffusion mechanism may control the drop vaporization, at least for these first five inches.

Further experimental work seems to be justified to determine more precise values of drop-size distribution, initial drop velocities, and liquid diffusion coefficients. Recalculation with the more correct values could possibly result in better agreement between the calculated and experimental values and give additional support to the proposed vaporization mechanism.

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NOTATION

A = area
C_p = heat capacity at constant pressure
d = drop diameter
D = diffusion coefficient
f = friction factor
g = gravitational constant
h = heat transfer coefficient
k = thermal conductivity
L = length
m = mass
M = molecular weight
r = drop radius
t = temperature
Δt = temperature difference, air-to-drop surface
u = velocity
x = mole-fraction high boiler
α_L = liquid thermal diffusivity
θ = time
λ = latent heat of vaporization
ρ = density

Subscripts

a = air
b = boiling point
h = horizontal
L = liquid
m = mean
0 = initial
v = vertical
HB = high boiler
LB = low boiler

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Evolution of Gas from Liquids Flowing through Porous Media

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The production of oil and gas by pressure depletion involves both the formation of bubbles and the diffusion of gas from the liquid phase into these bubbles. Studies were undertaken to outline in detail the process of bubble formation when the driving force is small. This work shows that the formation of bubbles is a random process which can, however, be described by a simple probability distribution function. Also, calculations have been made to determine how fast gas will diffuse into uniformly distributed gas bubbles.

These results make it possible to describe the manner in which a gas phase is established during the pressure depletion process. In this process the rate of pressure decline is the most important factor influencing the total number of bubbles produced. Laboratory tests have substantiated the finding that in certain types of porous media the amount of oil recovered is sensitive to the number of bubbles formed.

Oil is produced from many petroleum reservoirs by a solution-gas-drive process. In this production mechanism the underground oil is pushed out and replaced by its own dissolved gas when pressure on the reservoir is reduced. A typical oil field for which this process is operative is characterized by constant reservoir volume and is usually discovered at a pressure above the bubble-point pressure of the crude oil. When well-bore pressure is reduced, oil at first is produced by virtue